

AN EPIDEMIOLOGICAL STUDY USING PASSIVE RADON MEASUREMENT BY LIQUID SCINTILLATION COUNTING

R. J. A. KAPPEL¹, GERT KELLER¹, LOTHAR KREIENBROCK² and R. M. NICKELS¹

ABSTRACT. Radon (²²²Rn) and Rn daughters (²¹⁸Po, ²¹⁴Po) have been associated with the onset of lung cancer. The general public may be exposed to heavy Rn concentrations in indoor air. Thus, we have undertaken simple measures for determining Rn concentrations in air. We determined Rn concentrations in air by liquid scintillation counting (LSC) and analysis of exposure time, Rn decay, calibration factors and other parameters. We also compared this detection method successfully with another technique, using larger charcoal canisters and γ spectrometry, and with well-calibrated solid-state nuclear track detectors (SSNTD). We used our plastic LS vials for short-term measurements and SSNTDs for long-term measurements in an epidemiologic study of health risks of indoor Rn.

INTRODUCTION

Several epidemiological investigations of underground miners showed increased risk of Rn-induced lung cancer (BEIR IV), but differences in exposure conditions compared with normal living habits have not allowed direct comparison with the local population. For this reason, an epidemiological study is being conducted in Germany since 1989, in which indoor Rn concentrations and living habits of ~5000 individuals with diagnosed lung cancer (cases) and ~5000 healthy individuals (controls) were investigated. We describe our study of short-term Rn detection, using liquid scintillation counting (LSC).

METHODS

We used three passive methods to measure indoor Rn concentrations: two types of activated charcoal detectors for short-term measurements (2 or 3 days) and solid state nuclear track detectors (SSNTD) for long-term measurements (up to one year). The short-term measurements provide a rough estimate of the mean Rn concentration, and are necessary to optimize the exposure time of the SSNTDs.

The first type of charcoal detector is a special LS vial with a small, porous cartridge fixed in the upper half of the vial (Packard Picorad). The cartridge is filled with ~3 g activated charcoal mixed with a desiccant. The vial is capped by an air-tight (Rn-tight) lid. The desiccant ensures that the Rn adsorption is not (or minimally) influenced by moisture in the measured air.

The second type of detector for short-term measurements is a larger canister with ~70 g activated charcoal (EPA-type without diffusion barrier). The Rn concentration is determined by gamma spectroscopy with a NaI(Tl) detector. By making special corrections that affect the calibration factor, we accounted for the fact that the charcoal adsorbs macroscopic amounts of water during exposure (normally a few grams). Except for the efficiency of the γ detector, all corrections for the evaluation of the Rn concentration are given by EPA method 520/5-87-005 (EPA 1987).

The detector for long-term measurements is an aluminized polycarbonate foil (MAKROFOL™) inside a capsule (KfK-type) (Urban, Wicke & Kiefer 1984). After exposure, the microscopic damages in the foil (latent alpha tracks) were enlarged by an electrochemical etching process (ECE) to macroscopic dimensions. The etched tracks were automatically counted to determine the

¹Universität des Saarlandes, Institut für Biophysik, DW-6650 Homburg/Saar, Germany

²Bergische Universität GH Wuppertal, Arbeitssicherheit und Umweltmedizin, DW-5600 Wuppertal 1, Germany

exposure and the Rn concentration. Results of these long-term measurements are the main source for further epidemiological evaluations.

Normally, an LS vial was exposed for two days. Uncapping the vial allowed Rn gas to enter the cartridge and become adsorbed by the activated charcoal. The exact time of exposure must be known because the adsorption is a time-dependent process. The rate of Rn accumulation in the detector was determined empirically. After exposure, we added ~10 ml of a xylene-based LS cocktail to the recapped vial. The cocktail filled only the lower half of the vial. Because of its high vapor pressure, the cocktail desorbed the Rn from the charcoal only through its vapor phase. This prevented quenching effects because the cocktail was not soiled by activated charcoal. Figure 1 illustrates the course of the desorption process.

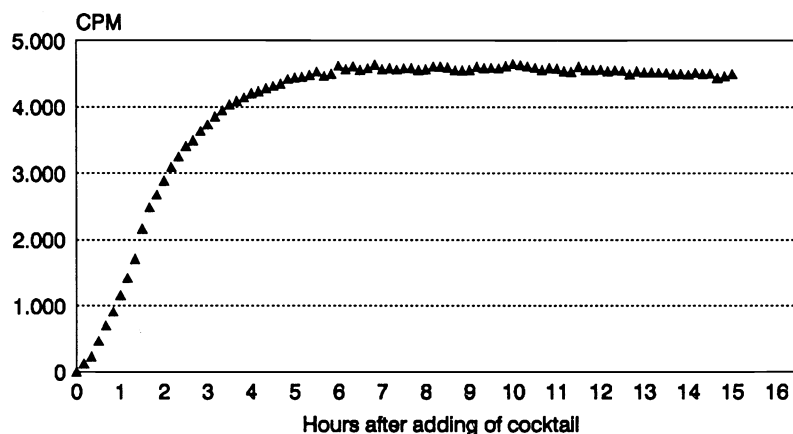


Fig. 1. Desorption of Rn after the cocktail is added

The maximum count rate is reached about 6–8 h after adding the cocktail. Then, Rn decay (half-life = 3.84 d) decreases the count rate.

DISCUSSION

The time between adding cocktail and beginning LS measurement normally exceeded 8 h; thus, a nearly complete desorption is ensured. (Any deviations in this time period is taken into account by the analysis program.) We evaluated Rn concentration by considering exposure time, decay time (time between recapping of the vial and start of measurement), empirical calibration factor and LS counter background. The LS counter uses the 25–900 keV energy region in the α - β spectrum, as shown in Figure 2. Table 1 lists ^{222}Rn , ^{218}Po and ^{214}Po α peaks.

We performed several tests under various conditions to ensure the comparability of different detection methods. The short-term charcoal canister and Picorad detectors were exposed at 14 locations (living rooms, bedrooms and basements), with Rn concentrations up to $25,000 \text{ Bq m}^{-3}$; three of each detector type, were employed at each sampling location. Figure 3 shows results of these measurements. The standard deviation of the results for both detector types was <10% with no significant difference between each type. However, absolute Rn concentrations differed, unpredictably. Exposures of both types of detectors together with well-calibrated SSNTDs in a Rn chamber at a nearly constant Rn concentration slightly modified results for the charcoal canisters.

TABLE 1. Alpha-Energies of Radon and Its Short-Lived Decay Products

Nuclide	α Energy (MeV)
^{222}Rn	5.486
^{218}Po	6.000
^{214}Po	7.688

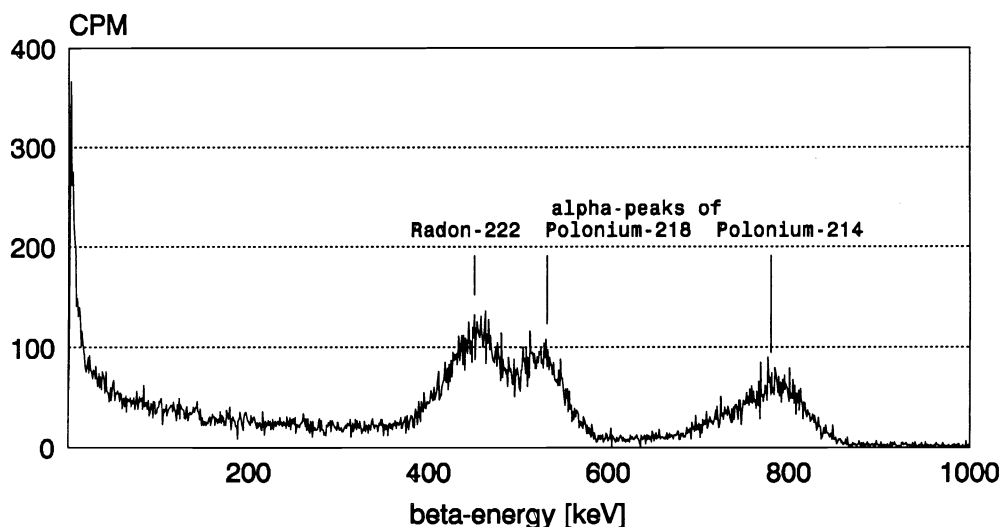


Fig. 2. Spectrum of the liquid scintillation analyzer

(The SSNTDs proved their reliability in various international calibration intercomparisons. Also, they are not influenced by high humidity, which has the greatest detrimental effect on charcoal measurements.) Recalibration was not necessary for LS vials because these results corresponded well with the SSNTD data.

PRELIMINARY RESULTS

During our case-control study, several thousand Rn measurements will be made in homes where individuals developed lung cancer as well as in the homes of control individuals. To date, we have made ~3500 short-term measurements and ~1500 long-term measurements. We made 500 short-term measurements with LS vials since February 1992. Figure 4 shows a frequency distribution of results exceeding the lower detection limit (15 Bq m^{-3}). Because seasonal variations (*e.g.*, increased room ventilation in spring and summer) reduce greatly indoor Rn concentrations, the mean Rn concentration is relatively low (Keller, Folkerts & Muth 1982). For this study, long-term measurements are more important because they should produce more accurate values of the annual Rn concentration (mean concentration of 1500 measurements = 69 Bq m^{-3} ; median concentration = 37 Bq m^{-3}).

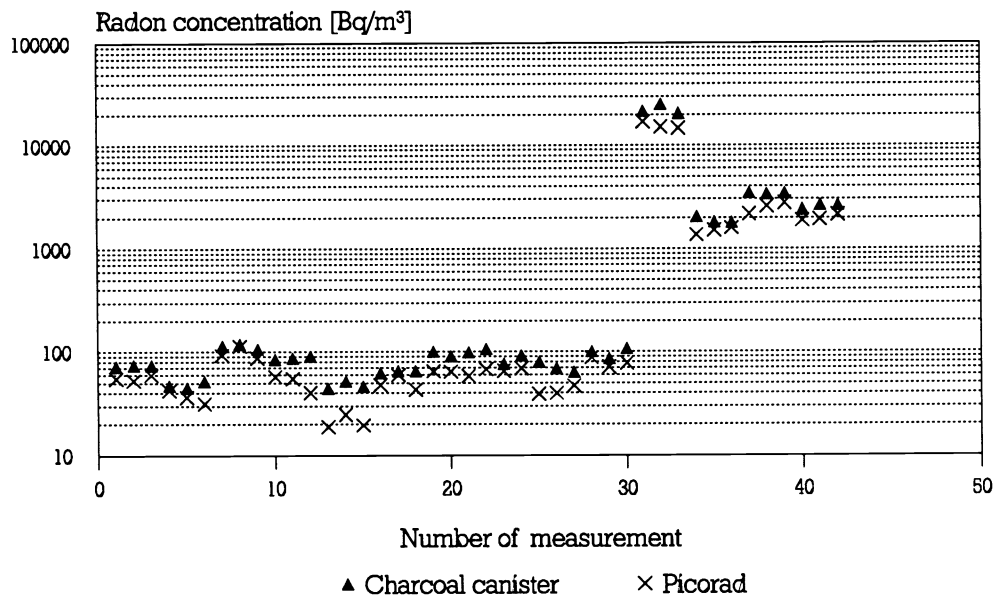


Fig. 3. Comparison of measurements with EPA charcoal canister and Picorad detectors (3 consecutive measurements each at 1 detector)

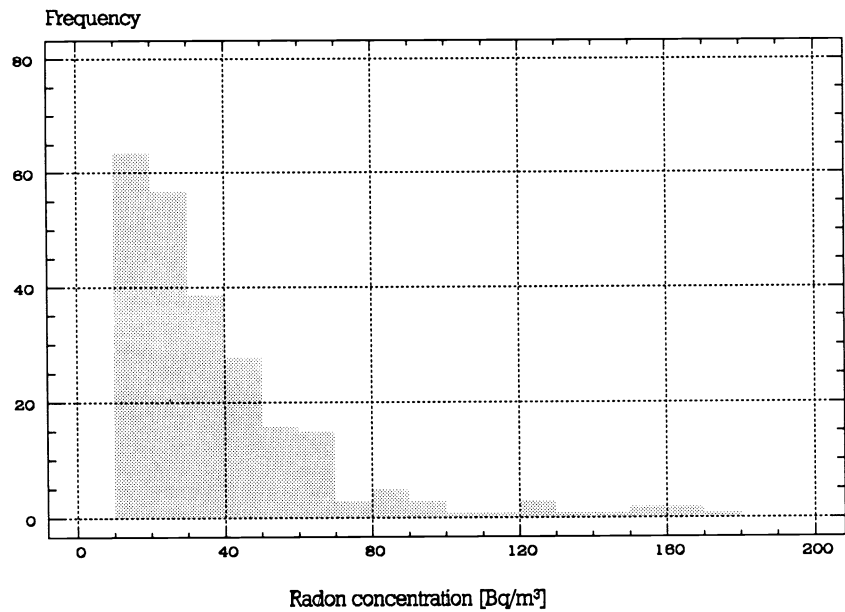


Fig. 4. Frequency distribution of ~250 Rn measurements with LS vials (Rn concentration = >15 Bq m⁻³); average = 45 Bq m⁻³; Median = 30 Bq m⁻³

CONCLUSION

Our results show that the LS method is well-suited for short-term measurements of indoor Rn concentration. Comparison with other types of short-term detectors show no great differences in accuracy. For the epidemiological study, both devices provide a rough estimate of long-term Rn concentration, which is used to determine the exposure time of SSNTDs.

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III. ENVIRONMENTAL APPLICATIONS

A. Tritium

B. Radon and Radium

C. Other Environmental Applications

